TR-1148

A STUDY OF TARGETS FOR MACHINE PRODUCTION OF 14-MEV NEUTRONS

Philip Berman

25 June 1963

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ABSTRACT

A study was made of available target materials suitable for production of an intense pulse of 14-Mev neutrons by the $D(T,n)He^4$ reaction. A basic requirement of the target is that it be inexpensive enough to be expendable. The target types studied were clathrate, zeolite, tritiated organics, and metallic tritides. The study indicates that synthetic zeolite in which tritium oxide (T_2O) is absorbed would provide the required yield and would be inexpensive enough to be expendable. Experimental studies of such a target are recommended.

1. INTRODUCTION

Theoretical studies into the mechanics of irradiation damage of solids indicate that damage is a direct function of neutron energy (ref 1, 2, 3) However, a quantitative measure of the damage is desirable. Therefore, data are required on which to base calculations. To provide such data, experiments using an intense fast-neutron beam of narrow spectrum is necessary. Freedom from gamma radiation is convenient but not a requirement. A brief survey was made by the author to determine the most practical and inexpensive means of providing such a beam. An accelerator appeared to be the most satisfactory source since nuclear detonations are limited by politics and geography and reactors produce a wide spectrum of neutron energies and quite frequently a large thermal neutron component. By basing the production of neutrons on the appropriate nuclear reaction and by controlling accelerator beam current and voltage, gamma-free neutrons with a narrow fast spectrum can be produced.

The D, T reaction is one of several nuclear reactions available (appendix) for the production of neutrons in an accelerator and was chosen for study since the spectrum is narrow and fast ($\sim$ 14 Mev) for a particular source-experiment geometry. The exact D, T reaction is $T + D \rightarrow He^4 + n + 17.6$ Mev, with a maximum cross section of 4.2 barns occurring at about 100 kev. Careful measurement of the neutron yield at various incident particle energies (acceleration voltages) has yielded cross-section data that satisfies the following equation (ref 4):

$$\sigma\,(E_{Mev}) = \frac{58}{E}\,\frac{e^{-1.72/E^{1/2}}}{1 + (E - 0.096)^2/(0.174)^2} \quad \text{barns}$$

The D, T reaction is a highly exoergic one. Therefore, the number of neutrons being emitted at a particular angle may be determined by counting the He^4 particles (α particles) associated with the neutrons, since these particles are rigorously associated with the direction of the neutrons through conservation of momentum properties. Thus, one does not have to detect high-energy neutrons.

The D, T reaction can be carried out by accelerating either deuterons or tritons into the appropriate target. The target may be either thick or thin, depending on the incident particle energies used. For thick targets, incident particle energies of at least 0.6 Mev should be used to take advantage of the large cross section throughout the entire resonance width. That is, incident particles should slow down in the target through the energy of maximum cross section (fig. 1).

Although variation of the energy of the incident particles governs the spread of neutron energies (ref 4), a variation of energies from 100 kev to 1 or more Mev is usable. Thus, many different types of machines may be used to generate high-energy neutrons. Cockcroft-Walton machines, Van de Graff machines, linear accelerators, and cyclotrons can be used. The greatest number of neutrons obviously would be produced by the machine with the largest beam current.

When the characteristics of the tritium targets are examined, problems of cost, as well as physics and chemistry, are found. Fabrication costs, availability, specific activity, and thickness combine to prevent the use of large beam currents with high incident particle energies on present targets, the most common of which is tritiated titanium. Metal targets tend to embrittle as well as degrade at high bombardment rates. Heating tends to cause the tritium to desorb from the metal backing. Also, if they are highly tritiated, they are expensive.

However, if a target were available that was cheap enough to be expendable and could be readily made to a thickness great enough to take advantage of the resonance width about 100 kev, a very intense source of 14-Mev neutrons would be available. In addition to being expendable, the target must be capable of being safely handled without elaborate equipment and easily made or readily purchased.

Three types of targets in addition to the metal tritide are considered -- a clathrate, a zeolite, and a tritiated organic compound. These targets are evaluated with respect to method of preparation, cost of tritium atoms, specific activity, and cost of equivalent macroscopic cross section.

2. EVALUATION

2.1 Clathrate

General Target Description--Clathrates are organic compounds having a crystal structure in which small molecules of another material, often a gas, are entrapped. The entrapped molecule must be small enough to fit into the crystal cage and too large to escape once the cage is completely formed Usually, entrapment of a gas is accomplished by crystallizing the clathrate from a solution pressurized by the gas. The quinones are the most widely used class of compounds for this purpose.

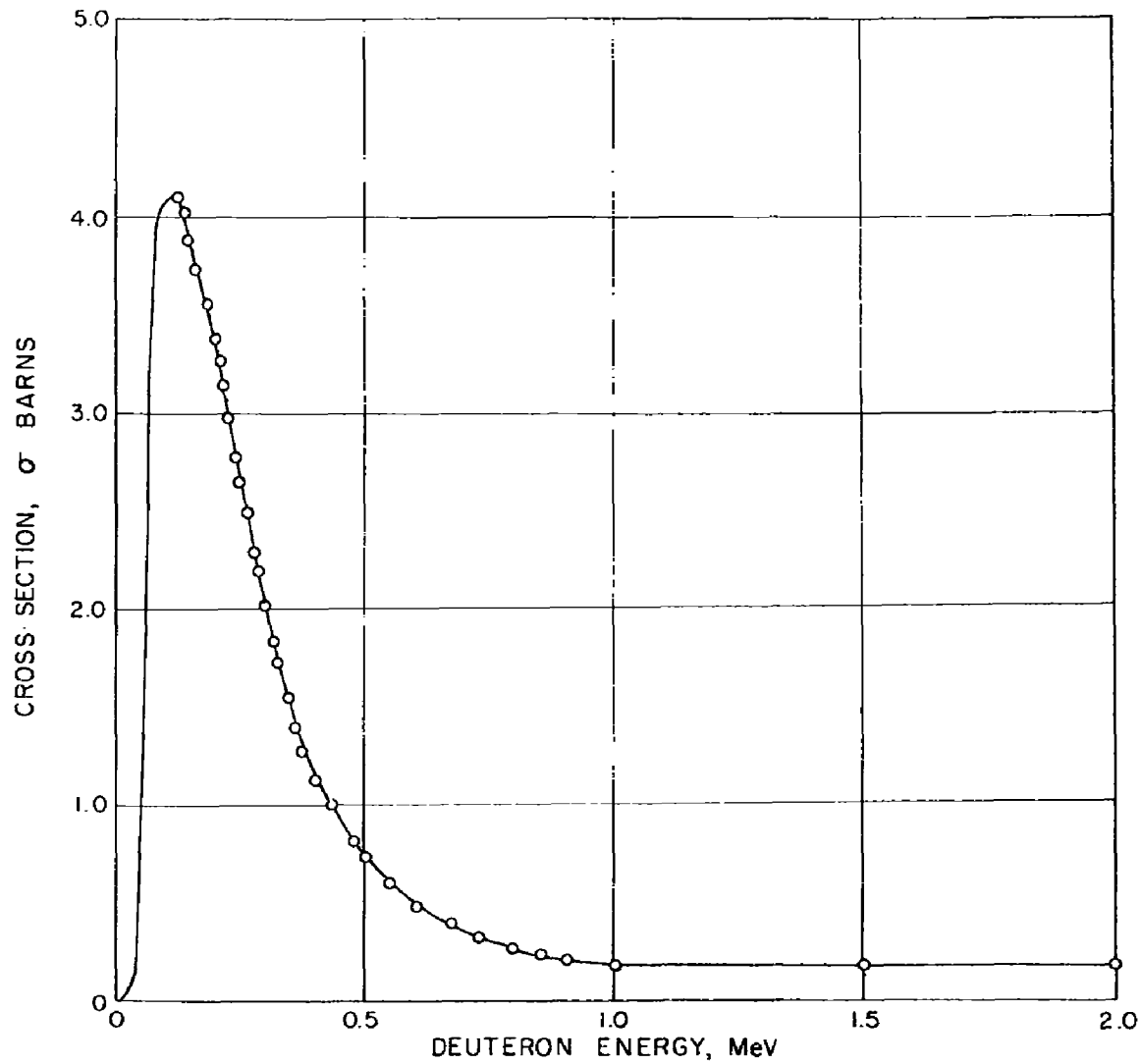

Figure 1. The total cross section of the $D(T,n)He^4$ reaction as a function of the incident deuteron energy.

The tritium molecule is too small to be entrapped into a clathrate; in the form of methane or ethane, however, a clathrate could be formed Use of tritium in the form of methane or ethane and subsequent entrapment, of course, increases cost of the target.

Specific Activity of Methane—A simple calculation shows that methane can theoretically be prepared with an initial activity of 116 curies per millimole of gas:

$$C = \left(\frac{1}{3.7\times10^{10}}\right)\left(\frac{dn}{dt}\right) = \frac{N(0.693)\ n'}{(T_{1/2})\ (3.7)(10^{10})}$$

$$C = \frac{(6\ 02)(10^{23})(4)(0.693)}{(12.3)(365)(24)(3600)(3.7)(10^{10})}$$

$$C = 1.163 \times 10^{2} \text{ curies per millimole}$$

where

$\frac{dn}{dt}$ = decay rate, disintegrations/sec

n' = number of tritiums possible per methane molecule

N = number of atoms/millimole

$T_{\frac{1}{2}}$ = half-life (years)

(365)(24)(3600) converts time from years to seconds

$(3.7)(10^{10})$ = disintegrations per second-curie

C = specific activity (curies per millimole)

With such a high level of activity, however, radiation degradation would quickly break down the methane molecules, possibly permitting the release of radioactive species. Also, degradation of the quinone must be considered because the intense beta radiation present can break the quinone molecule down and destroy the cage effect. An upper limit for specific activity of 5 curies per millimole for both methane and ethane has been suggested by a vendor of these types of compounds.

Tritium Number Density—The concentration of tritium atoms per cubic inch of target is determined by drawing an analogy between the methane—quinone clathrate and a krypton-85—quinone clathrate (ref 5). The specific activity obtained from the krypton-85 clathrate was 3 curies per gram. The number of krypton-85 atoms per cubic inch is computed as follows:

$$\frac{dn}{dt} = \frac{(0.693)N}{T_{1/2}}$$

C

Insertion of the values in compatible units yields

$$N = 1.14 \times 10^{21} \text{ krypton-85 atoms per cubic inch.}$$

If tritiated methane gas with a specific activity of 5 curies per millimole is considered, the average number of tritium atoms per molecule is calculated as follows:

$$N = \frac{dn}{dt} \frac{T_{1/2}}{0.693}$$

Insertion of the values in compatible units yields

$$N = 1.036 \times 10^{20} \text{ tritium atoms per millimole}$$

Then, by analogy, the tritium number density is

$$\frac{(1.036)(10^{20})}{(6.02)(10^{20})} = 0.17 \text{ tritium atoms per molecule of methane}$$

$$(0.17)(1.14)(10^{21}) = 0.194 \times 10^{21} \text{ tritium atoms per cubic inch of clathrate}$$

Target Specific Activity—Since tritium has a half-life of 12.3 yr, and the number of disintegrations per second per curie is 3.7×10^{10}, the specific activity in curies per cubic inch is computed as follows:

$$\frac{dn}{dt} = \frac{N\,(0.693)}{T_{1/2}}$$

$$C = \frac{19.4 \times 10^{18}}{3.7 \times 10^{10}} \quad \frac{0.693}{3.88 \times 10^{8}}$$

$$C = 9.36 \text{ curies per cubic inch}$$

Calculation of Cost—Hydroquinone, CP grade, from commercial sources, costs $45.40 for 20 pounds. Tritiated methane is commercially quoted at $2000.00 for 10 curies of a material with a specific activity of 5 curies per millimole. Since the specific activity of the methane clathrate is 9.36 curies per cubic inch, the cost of methane in the clathrate is $1876.00 per cubic inch. The cost of the hydroquinone is $0.11 per cubic inch.

2.2 Zeolite

General Target Description—Zeolites are considered primarily because they are inorganic, and therefore are not so prone to radiation degradation as the organic materials. Zeolites are commercially available being known as molecular sieves. Target preparation would

be simple. The zeolite material would be mounted according to the needs of the accelerator being used. The mounted zeolite would then be placed in a closed system where the tritium gas would be burned in dry air or dry oxygen. The resulting T_2O vapor would condense on and be absorbed by the mounted zeolite yielding a completely active, ready-to-use target. Since the targets could be easily prepared, storage would be unnecessary. Once the target is in the accelerator, any vapor released because of heating within the target during the pulse of the machine could be easily removed by using a drying agent.

Since the zeolite would be mounted on a backing prior to target activation, mechanical operations on a radioactive material would be avoided and the time lag between target activation and target utilization would be minimized. Removal of T_2O from the zeolite under normal conditions would be very difficult.

Tritium Number Density—For a representative zeolite, the following applies:

Hydrated density = 1.99 grams per cubic centimeter
Dehydrated density = 1.55 grams per cubic centimeter

At ordinary temperatures, a cubic centimeter of zeolite can contain 0.44 gram of water.

One hundred percent tritiated water can be prepared as described above, providing two atoms of tritium for each oxygen atom. The amount of radiation degradation that would occur is negligible due to the nature of the bond.

Since there are 22 grams of tritium oxide per mole, there could be 0.44/22 = 0.02 mole of tritium oxide per cubic centimeter of zeolite, or 0.02/0.061 = 0.32 mole of tritium oxide per cubic inch of zeolite. Since desorption of tritium oxide must be prevented, the zeolite must not be saturated. Therefore, the tritium oxide concentration might be limited to 0.02 mole per cubic inch of zeolite.

The number of atoms of tritium per cubic inch K is calculated as follows:

$$K = NM\,(6.02)(10^{23})$$
$$K = (2)(0.02)(6.02)(10^{23})$$
$$K = 0.24 \times 10^{23} \text{ tritium atoms per cubic inch}$$

where

N = number of tritium atoms per tritium oxide molecule
M = number of moles of tritium oxide per cubic inch of zeolite

Assuming that 0.02 mole of tritium oxide are entrapped per cubic inch of zeolite, a concentration of 2.4×10^{23} tritium atoms per cubic inch of zeolite is obtained.

Target Specific Activity--The specific activity of the target is computed as follows:

$$C = \frac{(2.44)(10^{22})}{(3.7)(10^{10})} \frac{(0.693)}{(3.88)(10^{8})}$$

$$C = 1.17 \times 10^{3} \text{ curies per cubic inch}$$

Cost--Carrier-free tritium can be purchased from Oak Ridge National Laboratories for $200 per curie plus a $30.00 flat rate per shipment for packing. With a specific activity of 1.17×10^{3} curies per cubic inch, the cost of the zeolite target would be $2340 per cubic inch. Cost of the dehydrated zeolite is about $0.30 per cubic inch.

2.3 Tritiated Plastics

General Description--Plastics offer interesting possibilities as accelerator target materials particularly since the tritium is chemically tied to the support material. Tritiated styrene has been commercially prepared to a specific activity of 32 curies per millimole. The styrene at such a high specific activity readily underwent radiation polymerization yielding a solid material. The tritiated polystyrene dissolves in benzene, and a thin-film target is easily obtained by putting a few drops of benzene-styrene solution on a plate and allowing the benzene to evaporate. Unfortunately, specific activities much in excess of 32 curies per millimole are not possible since radiation degradation of the polystyrene becomes excessive with attendant gas evolution.

Tritium Number Density--The number of tritium atoms per molecule of styrene monomer is

$$N = \frac{(32)(3.7)(10^{10})(3.88)(10^{8})}{0.693} = 6.63 \times 10^{3} \text{ tritium atoms per millimole}$$

$$= \frac{6.63 \times 10^{20}}{6.02 \times 10^{20}} = 1.1 \text{ tritium atoms per molecule of styrene monomer}$$

The molecular weight of styrene monomer is

$$(12.004)(8) + (6.9)(1.008) + (1.1)(3.017) = 106.36 \text{ grams per mole}$$

The number of tritium atoms per cubic inch of monomer is

$$\frac{(6.02)(10^{23})(0.902)(1.1)(0.061)}{106.36} = 3.43 \times 10^{20} \text{ tritium atoms per cubic inch of monomer}$$

Then by using the ratio of the densities of monomer to polymer, the tritium number density is

$$\frac{(1.06)(3.43)(10^{20})}{(0.902)} = 4.030 \times 10^{20} \text{ tritium atoms per cubic inch of polystyrene}$$

Cost—The cost of the tritiated polystyrene commercially is about $520.00 per curie. Therefore, the cost of target material is about $2,668,554 per cubic inch.

2.4 Metallic Tritides

General Description—At one time, zirconium tritide with a tritium-zirconium ratio of about 1:1 was preferred as an accelerator target. However, titanium is now much preferred. Tritiated titanium targets with a titanium-tritium atomic ratio of about 1:1 are commercially available and are in routine use. They are easy to work with but are difficult to prepare. They are usually purchased prefabricated and in the activated state.

A representative titanium tritide target is mounted on a stainless steel backing cut to either a square or rectangular shape. The thickness of the titanium tritide is about 0.7 micron.

Specific Activity—The representative target has a specific activity of about 1 curie per square inch of target. Thus, the specific activity of the titanium tritide is

$$\frac{1}{(0.7)(3.9)(10^{-5})(1)} = 3.66 \times 10^{4} \text{ curies per cubic inch}$$

Tritium Number Density—The tritium number density is

$$\frac{(3.66)(10^{4})(3.7)(10^{10})(12.3)(365)(24)(3600)}{(0.693)} = 7.6 \times 10^{23}$$

tritium atoms per cubic inch of titanium target

Cost—The representative target commercially costs $35.00 per square inch. The cost of the titanium tritide, then, is

$$\frac{\$35.00}{(2.75)(10^{-5})} = \$1.25 \times 10^{6} \text{ per cubic inch,}$$

where 2.75×10^{-5} is the volume of a square inch of target material on the target.

3. COMPARISON OF TARGET TYPES

3.1 Cost

The relative cost of the target types is shown in table I. The last column was calculated based on the following assumptions:

(a) One square inch of active target area is required.
(b) Each target could be used for one pulse only, since thermal effects would destroy it.
(c) Target thickness was determined to be that thickness of matrix material which would equal the deuteron range, neglecting tritium in the matrix.
(d) A 0.5-ma beam current was postulated for calculational purposes.
(e) Cost is normalized to 10^{10} neutrons per square centimeter per pulse-target

The data shows that the zeolite is far superior, costwise, to the other target types under consideration.

The basis for the results is shown in table II, where macroscopic cross sections are tabulated.

Variation in cost figures found herein from those published by vendors is due to the target thickness considered. All target costs are calculated on the basis that the target is thick enough to completely stop incident deuterons with energies up to 1 Mev. Table I does not contain cost information for machine down time due to target replacement, target-chamber cleanup, pumping down time, etc.

Table I. Target Number Density and Target Cost

Target type	Tritium number density, atoms/in^3	Target cost \$/in^3	Tritium cost \$/mole	Target cost \$/10^{10} n/cm^2-pulse
clathrate	1.94×10^{20}	1.9×10^3	5.8×10^6*	\$32 12
zeolite	2.40×10^{22}	2.3×10^3	58.7×10^3*	0.07
plastic	$4\ 03 \times 10^{20}$	2.7×10^6	$3\ 99 \times 10^9$*	3089.15
metallic tritide	7.60×10^{23}	$3\ 2 \times 10^3$	1.02×10^6 **	171.08

* Cost of materials only

** Cost of completely fabricated target, ready for use.

3.2 Cross Section

The microscopic cross section for the D, T reaction varies with deuteron energy as shown in figure 1. The maximum cross section of 4.2 barns occurs at an energy of 100 kev. Above 1 Mev, the cross section levels off at about 0.2 barns.

The macroscopic cross section, which in part determines target utilization efficiencies, was computed per cubic inch for each type of target at two energies, for the maximum microscopic cross section and for high energies (cross section is fairly constant above 1 Mev). Results are given in table II.

Table II. Macroscopic Tritium Cross Section per Cubic Inch of Target

Material	Cross section (cm^{-1}) 100 kev	> 1 Mev
clathrate	2.371×10^{-4}	1.159×10^{-5}
zeolite	0.250	0.012
plastic	3.520×10^{-3}	1.719×10^{-4}
metallic tritide	0.749	0.037

An effect not included in table II is the effective reaction cross section through the width of the resonance peak, as shown in figure 1. Utilization of the additional cross section thus available is obtained only through use of thick targets.

3.3 Target Preparation

Requirements for preparing the various types of targets are summarized in table III.

Table III. Requirements for Target Preparation

Material	
Clathrate	Facilities for fabricating tritiated methane or ethane. Apparatus for obtaining special conditions of temperature and pressure to form the clathrate.
zeolite	Closed system for burning tritium
plastic	Organic synthesis laboratory The process requires a relatively long time.
Metallic tritide	Metallurgical laboratory equipped to prepare and plate metallic tritides.

3.4 Target Integrity

All targets are certain to be destroyed by a large beam current of high-energy particles. A crude calculation considering a 15-ma beam current at 1 Mev gives a temperature rise of 10,050°C for 0.02 mole of water. For the zeolite target, the temperature rise would be much greater assuming 100-percent absorption of the beam in the target. Consequently, the target would lose its physical integrity releasing tritium oxide vapor to the target chamber volume. However, such vapor is easily removed by a desiccant in the vacuum line.

The amount of tritium oxide in the zeolite was chosen conservatively and arbitrarily. Experiments to determine the quantity of tritium oxide that zeolites can contain with no appreciable release at room temperature would be required.

3.5 Other Targets

The possibility of using highly tritiated organics as a target should be considered. Although water is an easily recaptured vapor compared with some organic vapors, proper selection of an organic compound would prevent the possibility of absorption of tritium oxide by personnel. However, the specific activity level would probably be greatly decreased relative to a tritium oxide target.

4. SUMMARY

A study was conducted to determine the suitability and availability of tritium-bearing targets to be used, with an accelerator, as a source of an intense pulse of fast neutrons for verification of quantitative theories relating to fast neutron damage in solids.

Since an intense pulse of neutrons is required, a target that is either cheap and expendable or capable of withstanding large temperature variations is needed. The assumption that a cheap one-shot target is better than an expensive, multishot target has been made. Thus, complete target destruction is assumed to occur for each pulse of neutrons produced.

Targets composed of tritium either chemically or physically bound to base materials of both organic and inorganic nature were considered. Conclusions are that an inorganic zeolite base would produce the cheapest target most easily. A fluence* of about 10^{10} n/cm^2 per pulse could be expected, 1 cm from the target.

An accelerator with a positive ion current capability of about 10 to 15 ma per pulse would be desirable. A series of experiments would be required to optimize neutron production in which accelerator voltage and target thickness are varied.

* Fluence is the term proposed by ICRU Report 10A for nvt. It is explained in NBS Handbook 84, 1962.

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Appendix Available Neutron-Yielding Reactions

Reaction	Half-life of target nucleus	Q value (Mev)	Neutron Energy (Mev)
$D(d,n)He^3$	stable	3.28	2.5
$N^{14}(d,n)O^{15}$	stable	5.1	4.7
$N^{15}(d,n)O^{16}$	stable	10	9.4
$T(d,n)He^4$	12.4 yr	17.6	14
$C^{12}(d,n)N^{13}$	stable	-0.26	0.005-2
$T(P,n)He^3$	12.4 yr	-0.764	0.060-3
$V^{51}(P,n)Cr^{51}$	stable	-1.5	0.4 energy near threshold
$Li^7(P,n)Be^7$	stable	-1.647	0.029-1.9
$Sc^{45}(P,n)Ti^{45}$	stable	-2.8	0.030 energy near threshold

Note that the last five reactions are endoergic and yield only low-energy neutrons. The first four reactions yield neutrons of relatively high energy as the energies indicated are the average energy of neutrons yielded by the reaction.

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2. Neutron beams—Generators

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